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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re the application of:

Hideyuki SAKAMOTO, et al.

Group Art Unit: 1713

Serial Number: 10/750,833

Examiner: Peter D Mulcahy

Filed: January 5, 2004

For: (METH)ACRYLIC RESIN COMPOSITION

DECLARATION UNDER 37 CFR 1.132Commissioner for Patents  
Washington, D.C. 20231

Sir:

Takahiko Sugaya residing at 4-10-34, Okihama-cho, Takasago-cho, Takasago-shi, Hyogo, 676-0026, Japan duly deposes and says:

1. That he graduated from Department of Synthetic Chemistry, School of Engineering Science, OSAKA UNIVERSITY, Osaka, Japan, in the year 1992, and he received the degree of Master of Chemistry from MASTER'S COURSE OF GRADUATED SCHOOL OF OSAKA UNIVERSITY, Osaka, Japan in the year 1994;
2. That since 1994, he has been employed in the capacity of KANEKA CORPORATION.;
3. That from 1994 he has been engaged in development for resin modifiers.;
4. That he has read and is familiar with the instant application for United States Letters Patent and Office Action thereto mailed on June 1, 2006; and

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5. That he has made experiments in order to show that the present invention can provide a (meth)acrylic resin composition, wherein heat degradation is decreased considerably, by the combination of a copolymer obtained by suspension polymerization and a fatty acid metallic salt.

### Example 3

#### <Preparation of suspension polymer>

A reaction vessel equipped with a stirrer was charged with 220 parts of deionized water and 15 parts by weight of a 3 % aqueous solution of polyvinyl alcohol (GH-20: available from The Nippon Synthetic Chemical Industry Co., Ltd.) and the inside of the reaction vessel was substituted with nitrogen. Then, 1 part by weight of lauroyl peroxide was dissolved into a monomer mixture comprising 90 parts by weight of methyl methacrylate and 10 parts by weight of butyl acrylate, and the rotational speed of the stirrer was adjusted so that the dispersion particle size of the monomer became approximately 250  $\mu\text{m}$ . Thereafter, polymerization was completed by raising the temperature in steps and heating at 60°C for 2 hours, 70°C for 2 hours, 80°C for 2 hours and 90°C for 1 hour, to prepare a suspension polymer having a polymer solid content concentration of 30 % by weight. The obtained polymer was washed and dried by a known method to obtain suspension polymer particles in the form of beads.

#### <Preparation of graft copolymer having a multi-layer structure>

##### (a) Polymerization of innermost layer

A glass reaction vessel was charged with a mixture of the

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following composition and the temperature was raised to 80°C while stirring in nitrogen current. Then, 25 % of the mixture of the innermost layer components comprising 24 parts by weight of methyl methacrylate, 1 part by weight of butyl acrylate, 0.1 part by weight of allyl methacrylate and 0.1 part by weight of t-butyl hydroperoxide was added at once and polymerization was conducted for 45 minutes.

Mixture:	(parts)
Deionized water	220
Boric acid	0.3
Sodium carbonate	0.03
N-lauroyl sodium sarcosinate	0.09
Formaldehyde sodium sulfoxylate	0.09
Ethylenediamine sodium tetraacetate	0.006
Ferrous sulfate heptahydrate	0.002

Subsequently, the remaining 75 % of the mixture was continuously added over 1 hour. After adding, the temperature was maintained for 2 hours to complete polymerization. During this time, 0.2 part by weight of N-lauroyl sodium sarcosinate was added. The average particle size of the polymer particles in the latex of the obtained crosslinked methacrylic polymer was 1600 Å (found using light scattering of wavelength of 546 nm) and the polymerization conversion ratio ((amount of polymers produced/amount of monomers charged) × 100) was 98 %.

(b) Polymerization of rubbery polymer

A latex of the crosslinked methacrylic polymer obtained in (a) was maintained at 80°C in a nitrogen current. After 0.1 part by weight of potassium persulfate was added, a monomer mixture

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comprising 41 parts by weight of n-butyl acrylate, 9 parts by weight of styrene and 1 part by weight of allyl methacrylate was continuously added over 5 hours. During this time, 0.1 parts by weight of potassium oleate was added divided into three times. After adding the monomer mixture, 0.05 parts by weight of potassium persulfate was added and the temperature was maintained for 2 hours to complete polymerization. The average particle size of the obtained polymer was 2300 Å and the polymerization conversion ratio was 99 %.

(c) Polymerization of outermost layer

A latex of the rubbery polymer obtained in (b) was maintained at 80°C. After 0.02 part by weight of potassium persulfate was added, a mixture comprising 24 parts by weight of methyl methacrylate, 1 part by weight of n-butyl acrylate and 0.1 part by weight of t-dodecyl mercaptan was continuously added over 1 hour. After adding the monomer mixture, the temperature was maintained for 1 hour to obtain a latex of a three-layer structure graft copolymer. The average particle size of the obtained three-layer structure graft copolymer 2530 Å and the polymerization conversion ratio was 99 %. The latex of the obtained three-layer structure graft copolymer was salted-out and coagulated, thermally treated and dried by a known method to obtain a copolymer having a three-layer structure in the form of white powder (impact modifier).

<Composition of (meth)acrylic resin composition and preparation of molded article>

0.5 part by weight of SC-100 (calcium stearate, available from Sakai Chemical Industry Co., Ltd.), which is a fatty acid metallic

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salt, was added to 100 parts by weight of a mixture comprising 75 % by weight of the obtained suspension polymer and 25 % by weight of the graft copolymer having multi-layer structure. 40 g of the mixture were charged into the chamber equipped with LABO PLASTMILL MODEL 20C200, made by Toyo Seiki Seisaku-Sho, Ltd. (mixer: R60H, roller type blade, temperature: 240°C, rotation number of cokneader: 50 rpm) and were kneaded for 18 minutes. The kneaded mixture was brought out from the chamber soon after kneading, and pressed to obtain a plate with a thickness of 3 mm by using a press machine under conditions of 190°C, for 10 minutes. The plate was left for overnight at 23°C. Then, b value (the degree of yellowing) of the obtained plate was evaluated in accordance with JIS K7105 using a color-difference meter (made by Nippon Denshoku Industries Co., Ltd., ZE-2000 Color Measuring System). The results are shown in Table 1.

#### **Comparative Example 3**

The plate for evaluation was obtained in the same manner as in Example 3, except that the fatty acid metallic salt (calcium stearate) was not used.

#### **Comparative Example 4**

The plate for evaluation was obtained in the same manner as in Example 3, except that the ACPE629A (oxidized polyethylene wax, available from Allied Signal Inc.) was used instead of calcium stearate.

#### **Comparative Example 5**

The plate for evaluation was obtained in the same manner

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as in Example 3, 100 parts by weight of a mixture comprising 75 % by weight of the commercially available PMMA obtained by bulk polymerization (Acrylite® H-12, available from CYRO Industries) and 25 % by weight of the graft copolymer having multi-layer structure obtained in Example 3 were used instead of the mixture in Example 3.

#### **Comparative Example 6**

The plate for evaluation was obtained in the same manner as in Comparative Example 3, except that the fatty acid metallic salt (calcium stearate) was not used.

#### **Comparative Example 7**

The plate for evaluation was obtained in the same manner as in Comparative Example 3, except that the ACPE629A (oxidized polyethylene wax, available from Allied Signal Inc.) was used instead of calcium stearate.

The results of the above evaluations are shown in the following Table 1.

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Table 1

Example		Comparative Example				
3		3	4	5	6	7
Method for obtaining acrylic resin	S	S	S	B	B	B
	calcium stearate		polyethylene wax	calcium stearate		polyethylene wax
b value	20.4	29.0	31.4	14.9	7.6	13.0
S: Suspension polymerization						
B: Bulk polymerization						

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**Result and Discussion**

As evident from the results of Table 1, when a copolymer is obtained by bulk polymerization, even if a fatty acid metallic salt is used together (Comparative Example 5), b value is increased to 14.9 compared with Comparative Example 6, wherein no additives are used. Also, when oxidized polyethylene wax is used instead of a fatty acid metallic salt (Comparative Example 7), b value is increased to 13.0. Similar results are observed, when oxidized polyethylene wax or no additives are used in combination with the copolymer obtained by suspension polymerization (Comparative Example 4 and 3). In Comparative 4 and 3, b values are increased to 31.4 and 29.0, respectively, which are considerably high compared with Example 3 represented as follows.

In Example 3, when the copolymer obtained by suspension polymerization are used with a fatty acid metallic salt, b value can possess considerably low value of 20.4.

Consequently, it is clear that the present invention can improve heat degradation of the obtained (meth)acrylic resin only when the copolymer obtained by suspension polymerization and a fatty acid metallic salt are used together.



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The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 24th day of August, 2006

by Takahiko Sugaya  
Takahiko Sugaya

We, the undersigned witnesses, hereby acknowledge that Takahiko Sugaya is personally known to us and did execute the foregoing Declaration in our presence on:

Date: August 24, 2006      Witness Kakumasa Nakajima

Date: August 24, 2006      Witness Akio Sato